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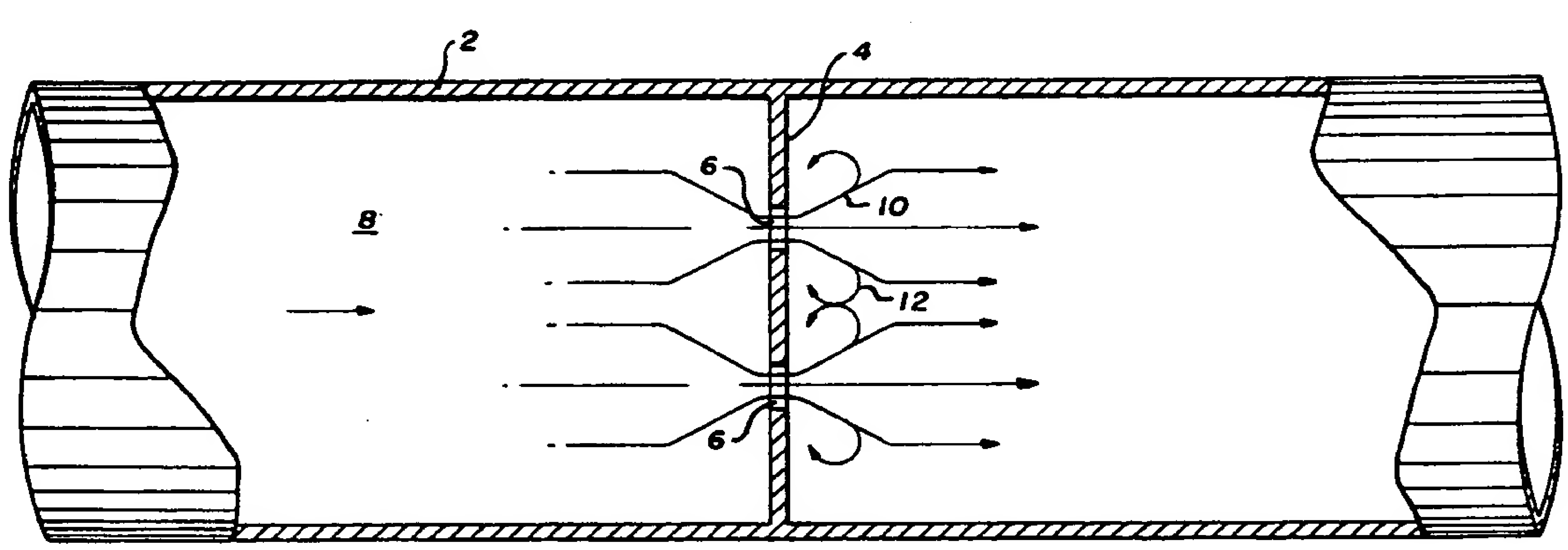
(54) **Jet impingement reactor.**

(57) An apparatus to allow reaction in the liquid phase. The apparatus is a vessel (2) having a baffle (4). There are openings (6) in the baffle (4) through each of which a liquid (8) passes as a jet (10). Neighboring openings (6) are sufficiently close to allow impingement of the jets (10) thereby allowing for reaction of the liquids. The invention finds application in reactions where the reactants are immiscible and is particularly suitable in the nitration of aromatic hydrocarbons using mixed acids in aqueous solution. A method of conducting a reaction between at least two reactants in the liquid phase is also provided comprising passing a liquid containing the reactants through a plurality of adjacent spaced openings to create a series of impinging jets.

Fig. 1.



EP 0 489 211 A1

This invention relates to an apparatus to allow a reaction in the liquid phase and to a method for conducting a reaction. The invention finds application in reactions where the reactants are immiscible. The invention is of particular application in the nitration of aromatic hydrocarbons using mixed acids in aqueous solution.

It is known that vigorous agitation is required for nitration reactions between an aromatic hydrocarbon and a mixture of sulfuric acid and nitric acid, commonly called mixed acid. Most of the known nitration processes using mixed acid use reactions vessels that incorporate agitation. These reactions are notoriously dangerous. They are highly exothermic and potentially explosive but it is well known that the risks inherent with these processes can be reduced if the charge of unreacted components can be made small.

It is also well known that the formation of undesirable by-products is increased as residence time within the apparatus is increased. For example, in processes for the manufacture of mononitrobenzene, United States Patent 4,021,498 to Alexanderson recognized that a reaction time of 0.5 to 3 minutes was preferred and United States Patent 2,256,999 to Castner indicates a complete reaction in about 10 minutes. It is not as well known that by-product production also increases with temperature.

It has, however, been found that when such processes are scaled up the efficiency of the reactant conversion is often less than that achieved on a small scale. This reduction in efficiency is commonly overcome by adding further conventional stirred tank reactors to the system. This has the effect of increasing the residence time, which increases the charge of unreacted and reacted components and increases the formation of undesirable by-products. Inevitably, the continuous stirred tank reactor, when operated in a manner necessary to provide the desired vigorous agitation, is subject to wear and mechanical breakdown.

United States Patent 4,453,027 to Vaidyanathan teaches that halobenzenes can be nitrated in a tubular reactor of the static-mixer type. It has been found, however, that the efficiency of these static-mixers is also reduced when scaled up to sizes practical for large scale production. This is probably due to the comparatively low velocities available within the constraints of space and residence time.

It is therefore recognized that a need exists for apparatus that permits nitration processes to operate efficiently and safely in large commercial applications.

Prior art devices for handling fluids are well known, however, these devices are generally limited to performing mixing and blending operations.

U.S. Patent 4,514,095 to Ehrfeld et al. discloses a motionless mixer in which a series of discs are arranged so that fluid passing through the mixer is divided into a number of streams whereupon the streams are recombined to thoroughly blend the fluid.

U.S. Patent 4,043,539 to Gilmer et al. teaches a static-type mixer comprising a conduit that separates a fluid or fluids to be mixed into a series of parallel streams. A portion of the fluid is diverted laterally from a main passage and the remainder of the flow is then reversed to rejoin the diverted portion in order to produce a mixing effect.

U.S. Patent 4,043,539 to Leffelman also teaches a static mixing device comprising a cylinder having an inlet and outlet and a plurality of hollow spheres with openings therethrough mounted within the cylinder. Fluids flowing through the cylinder are mixed in the turbulent flow that is created about the spheres.

U.S. Patent 4,361,407 to Pelligrini discloses a further example of a stationary mixing device that uses a series of separable stages in which are formed cavities and alignable holes to define passages for the flow of fluids to be mixed. Fluids are divided and recombined in the passages to create an essentially homogeneous mixture after passing through several of the stages.

The devices of the prior art are essentially concerned with mixing or blending of miscible fluids. In contrast, the apparatus and method of the present application is concerned with accelerating reactions between immiscible fluids that have been previously mixed. The apparatus of the present application accepts a flowing fluid comprising two or more immiscible and reactive liquids and uses the energy from the flow of the fluid to create a high shear on the fluid that breaks up a portion of the flow into small droplets having a large exposed surface area. These small droplets provide a greatly increased surface area for chemical reaction between the liquids thereby greatly accelerating the reaction rate. The shearing action is achieved by passing the fluid through sharp edges holes, and by impinging the resulting jets against a surface or against other jets or a slower moving fluid.

Accordingly, the present invention provides an apparatus to allow reaction in the liquid phase and comprising:

a vessel having a longitudinal axis; a baffle in the vessel;

a plurality of first openings in the baffle through each of which a liquid passes as a jet, neighboring openings being spaced to allow impingement of the jets.

In a further aspect the present invention is a method of conducting a reaction between at least two reactants in the liquid phase comprising:

passing a liquid containing the reactants through a plurality of adjacent spaced openings to create a series of impinging jets.

Aspects of the invention are illustrated, by way of example, in the accompanying drawings in which:

Figure 1 is a side elevation, partially in section, of an apparatus according to the present invention;

Figure 2 is a view similar to Figure 1 of a further embodiment of the present invention;

Figure 3 is a section through yet a further embodiment of the invention;

Figure 4 shows a development of the embodiment of Figure 3;

Figure 5 is a section of a further apparatus according to the present invention;

Figure 6 illustrates yet a further embodiment of the present invention;

Figure 7 shows an inlet system for introducing reactants into the apparatus of the present invention;

Figure 8 is a section view through the inlet system taken along line 8-8 of Figure 7;

Figure 9 is a side elevation, in section, of a further embodiment of the present invention; and

Figure 10 is a detail of a variation of the Figure 9 embodiment.

Each drawing shows an apparatus according to the present invention.

Figure 1 shows a reactor comprising a vessel 2 in the form of an open-ended cylinder. There is a baffle 4 in the vessel 2 and a plurality of first openings 6 in the baffle 4. Through each of these openings 6, a liquid 8, passing through the vessel 2, passes as a jet 10. The openings 6 are arranged sufficiently close to allow impingement of the jets 10, as schematically illustrated by the arrows 12 in Figure 1.

Figure 2 shows the presence of a second baffle 14, spaced downstream from the first baffle 4. There is a plurality of second openings 16 in the second baffle 14. The second openings 16 are arranged so that the first and second openings 6 and 16 are not aligned. Thus the jets 10 from the first openings impinge on the second baffle 14 as shown by the arrows 18 in Figure 2. There is an inlet for further reactants at 19 and further baffles, with openings, are placed downstream to provide a further reaction location.

In the embodiments of Figures 1 and 2, the first and second openings 6 and 16 are both arranged to direct the jets 10 longitudinally of the apparatus. In both cases, the first and second baffles 4 and 14 extend transversely of the vessel 2. However, Figure 3 illustrates an embodiment of the invention in which the baffle 20 comprises an annulus extending inwardly from the periphery of the vessel 2. A cylinder 22 extends longitudinally of the vessel 2, from the inner periphery of the annular baffle 20, to terminate in a closure 24 that is parallel to the annular baffle 20. Openings 26 are formed in the cylinder 22 so that jets 28 are directed by the openings 26 transverse of the vessel 2. In the embodiment of Figure 4 there is a plurality of cylinders 30, each having first openings 32, extending from annular walls 34 and 36. Again inlet 19 for further reactants is present in Figures 3 and 4 and there will be a further reaction location downstream.

Figure 5 illustrates an apparatus in which there is a plurality of generally coaxial cylinders 38, 40 and 42, each extending from an annular wall 44 extending from the periphery of vessel 2. Openings 44, 48 and 50 are arranged so that the liquid 8 flowing through an opening in an inner cylinder impinges on the wall of an outer cylinder before it can pass through openings in that outer cylinder.

Figure 5 also illustrates a particular embodiment of the invention in which there are opposed cylinders. Thus Figure 5 also shows cylinders 52, 54 and 56 extending from annular wall 58 towards wall 44. Openings 60, 62 and 64 are formed in cylinders 52, 54 and 56.

Figure 6 illustrates an embodiment of the invention in which baffles 66 are formed as generally concentric spheres 68, 70 and 72 each having inlet openings 74 and outlet openings 76 arranged so that liquid flowing in the vessel 2 must pass through the inlets 74, to the inner spheres 68, then outwardly.

Reactants can be added to the embodiment.of Figure 6 through inlet 19. An inlet system that uses a multiplicity of pipes distributed radially around reactor vessel 2 may be also be used. The location of the inlet pipes 19 may also be between stages of the concentric spheres, as shown in Figure 7. Figure 8 shows a sectional view through the multiple delivery pipes 19 to demonstrate the arrangement of the pipes through the vessel walls and into the concentric spheres. The number and size of the inlet pipes 19 are arranged to ensure a very high velocity jet, with very small droplets entering the reactor.

Figure 9 illustrates an embodiment of the invention resembling that of Figure 7 and common reference numerals are used where appropriate. However, Figure 9 also shows the use of semi-spherical baffles 78 arranged concentrically around the sphere 68.

The sphere 68 on the left of Figure 9 has two semi-spherical baffles. On the right of Figure 9 there are two spherical baffles 68 and 70 and one semi-spherical baffle 78, downstream of the pair of spherical baffles 68 and 70.

Figure 10 also shows the relationship of the embodiment of Figure 9 to the embodiment of Figure 7 in showing multiple inlet pipes 19 extending through the vessel walls and into the sphere 68.

Using the apparatus of the present invention the local velocity of each stage can be made sufficiently high to create conditions necessary for a nitration reaction between an aromatic hydrocarbon and mixed acid in the liquid 8 to take place independently from the bulk velocities of the reactants passing through the apparatus. The proportions of the apparatus can be adjusted, using simple experimental techniques, to achieve a wide range of intensive agitation and residence time.

The apparatus can be used either as a single unit or as a number of units connected in series or in conjunction with one or more continuously stirred tank reactors.

The apparatus of the invention is immediately of use in the adiabatic mononitration of benzene because of the large scale manufacture of this product. However, the invention can also be used in the nitration of other aromatic hydrocarbons or halogen substituted aromatic hydrocarbons.

The particular benefit provided by the present invention is the degree of agitation that is available. This ensures that the reaction rate and conversion efficiency of the reactor are high. The desired high agitation is accomplished by causing the jets containing the liquid 8 of aromatic hydrocarbon and mixed acids to be directed towards each other so as to provide varying degrees of impingement of the jets. This impingement, or interplay, of the jet produces high shear rates in the liquid, much higher for example than provided by propeller blades in a conventional stirred tank reactor or than of the shearing rates in a static mixer reactor. In addition to the shear between the jets a certain portion of the jet streams will directly impinge so as to bring droplets of the dispersed phase into direct contact and further enhance the reaction. The direct impingement of the jets, along with the relative shear between the jets, will produce a constant supply of fresh interface between the reacting components, thereby enhancing the reaction rate and overall conversion efficiency of the reactor.

An additional benefit provided by the present invention is the ability to add reactants in a high velocity jet directly into a region of high-intensity mixing as shown by the inlet system of Figures 7 and 8. The high velocity produces a jet of small droplets having a high surface area to mass ratio, thereby promoting the overall conversion of the reactants.

The particular arrangement used to bring about jet impingement will vary according to the rate of reaction required. In the simplest form, as shown in Figures 1 and 2, the lowest degree of impingement is provided. The liquid jets are disposed parallel. The impingement occurs when the jets spread and combine in a downstream direction. Impingement is due to lateral components of the turbulent velocity in the jets.

The embodiment of Figure 2, with its downstream impingement plate, causes the jets to change direction and impinge more directly. The provision of orifices in the second plate ensures a second stage of reaction. Further amounts of reactants can then be introduced through inlet 19 to increase the efficiency of conversion and minimize by-product formation. That is, still further stages or reaction locations can be arranged, depending on the degree of reaction required.

In the Figure 3 embodiment the jets are turned so that they impinge on the wall of the reactor. In this embodiment the impingement, shearing and mixing of the components is further enhanced by the requirement of the fluid to turn back into the main fluid direction, as shown by the arrows. Such an arrangement can also be repeated in stages to the desired degree of reaction.

In the embodiment of Figure 4, the multiplicity of lateral jets ensures that some of the liquid jets will impinge directly on each other, achieving the highest possible degree of agitation and therefore reaction rate. The arrangement of annular walls and cylinders shown in Figure 4 can be repeated downstream for further conversion, if required. Further reactants can be added through inlet 19 prior to each stage as discussed above for Figure 2.

Figures 1 to 4 show the flow direction to be axial, but the same principles can also be used if the flow arrangement be radial as shown by the cylindrical arrangement of Figure 5, and the spherical arrangement of Figure 6.

In Figure 5 the flow issues outwardly through a series of cylinders. The successive outward cylinders are preferably arranged so that the openings are not in line, producing the maximum benefit of reaction as discussed for Figure 2. The same arrangement can be used equally with the flow passing radially inwardly through the cylindrical shells. Again reactants may be added between the two stages through inlet 19. The first stage is defined by cylinders 38, 40 and 42 and the second by cylinders 52, 54 and 56. Again this reactant addition between stages improves conversion.

In Figure 6 the flow issues outward through a series of spheres, each with openings to produce jets. The openings are successively offset to produce maximum reactions as in the case of Figure 2. Flow can also be directed radially inward, that is opposite to that shown in Figure 6, and combination of radial inflow and outflow can be combined to form a compact stage. Many more stages can be added in the continuation of this principle.

In Figure 7, the reactants are introduced directly between the concentric spheres shown through a plurality of inlet pipes 19 arranged radially about vessel 2. The size and number of the inlets is chosen appropriately so that the reactant jet velocity is very high. This promotes the formation of small droplets of reactant which leads to high overall reaction rates and high conversion efficiency.

Figures 9 and 10 show that hemispheres may be used to achieve the same effect as spheres. Hemispheres may be arranged in any combination or number on the upstream or the downstream side of the spheres. The preferred arrangement depends on the degree of reaction desired and would be determined for any particular set of reaction conditions by routine experiment. Figure 10 shows that the arrangement that includes inlet pipes is also compatible with the semi-spherical arrangement shown in Figure 9.

A virtue of the apparatus according to the invention is compactness where the prior art equipment can be massive. Thus, in the embodiments of Figures 1 to 4, the vessels 2 can be cylinders of a diameter within the range 6 to 12 inches. The openings 6, 16, 26 and 32 may have a diameter of about 1/2 inch. They are symmetrically arranged in walls 4 and 14.

Flow rates can, for example, be in the range of 100 to 800 U.S. gallons per minute.

In the embodiment of Figures 5 and 6 the end pipes shown may, for example, have diameters of about 8 inches. The vessels 2 have, for example, diameters of about 12 inches. Openings 60, 62, 64, 74 and 76 have diameters, for example, in the range 1/4 to 1/2 inch.

In the embodiment of Figure 7, the inlet pipes 19 may be 1/16 to 5/16 inch with any number of such inlets, for example, 32, disposed radially about the reactor vessel. This embodiment could be used, for example, if only the aromatic hydrocarbon is being added through the inlets.

The apparatus may be made of glass lined steel, as in the prior art, but preferably are made from zirconium or tantalum or any suitable corrosion-resistant material.

Claims

1. Apparatus to allow reaction in the liquid phase and comprising:

   a vessel having a longitudinal axis; a baffle in the vessel;

   a plurality of first openings in the baffle through each of which a liquid passes as a jet, neighboring openings being spaced to allow impingement of the jets.

2. Apparatus as claimed in claim 1 including a second baffle;

   a plurality of second openings in the second baffle arranged so that the first and second openings are not aligned so that the jets from the first openings impinge on the second baffle.

3. Apparatus as claimed in claim 1 in which the first openings extend through the baffle parallel to the longitudinal axis of the vessel such that the openings direct the jets longitudinally of the apparatus.

4. Apparatus as claimed in claim 1 in which the baffle is a plate extending transversely of the longitudinal axis of the vessel.

5. Apparatus as claimed in claim 1 in which the first openings extend through the baffle at an angle to the longitudinal axis of the vessel such that the openings direct the jets transversely of the apparatus.

6. Apparatus as claimed in claim 5 in which said vessel has an outer wall and the baffle comprises an annular wall extending inwardly from the periphery of the outer wall, said annular wall having an inner peripheral edge;

   a cylindrical portion having longitudinal walls extending along the longitudinal axis of the vessel from the inner peripheral edge of the annular wall and terminating in a closure parallel to the annular wall, the first openings being formed in the longitudinal walls.

7. Apparatus as claimed in claim 6 having a plurality of cylindrical portions, each having first openings.

8. Apparatus as claimed in claim 6 having a plurality of generally coaxial cylindrical portions.

9. Apparatus as claimed in claim 7 having opposed cylindrical portions, each comprising a plurality of generally coaxial cylinders extending towards each other from remote annular walls.

10. Apparatus as claimed in claim 1 in which the baffle is generally spherical.

11. Apparatus as claimed in claim 10 having a plurality of spheres of different diameters, arranged one within the other, each sphere formed with a plurality of first openings acting as inlets and a second plurality of openings acting as outlets.

12. Apparatus as claimed in claim 1 including a plurality of stages, each stage comprising at least one baffle with a plurality of openings;
an inlet for further reactants between each stage.

13. Apparatus as claimed in claim 12 in which each stage comprises a plurality of baffles.

14. Apparatus as claimed in claim 1 including an inlet for introducing reactants,

15. Apparatus as claimed in claim 14 in which said inlet comprises a plurality of pipes arranged radially about said vessel.

16. Apparatus as claimed in claim 11 including an inlet for introducing reactants, said inlet comprising a plurality of pipes arranged radially and extending through said spheres.

17. Apparatus as claimed in claim 16 in which said inlet pipes open into the sphere having the smallest diameter.

18. Apparatus as claimed in claim 10 in which the generally spherical baffle has at least one semi-spherical baffle concentric with it.

19. Apparatus as claimed in claim 18 in which the semi-spherical baffle is upstream relative to the spherical baffle.

20. Apparatus as claimed in claim 18 including an inlet for introducing reactants, said inlet comprising a plurality of pipes arranged radially and extending into said sphere.

21. Apparatus as claimed in claim 11 in which the plurality of spheres has at least one semi-spherical baffle associated with it.

22. A method of conducting a reaction between at least two reactants in the liquid phase comprising:
passing a liquid containing the reactants through a plurality of adjacent spaced openings to create a series of impinging jets.

23. A method as claimed in claim 22 including introducing the reactants through inlet pipes arranged to produce a high velocity jet of small droplets.

24. A method as claimed in claim 23 in which only some reactants are introduced through inlet pipes.

25. A method as claimed in claim 22 in which the reactants are immiscible.

26. A method as claimed in claim 22 in which the reaction is nitration and one of the reactants is nitric acid.

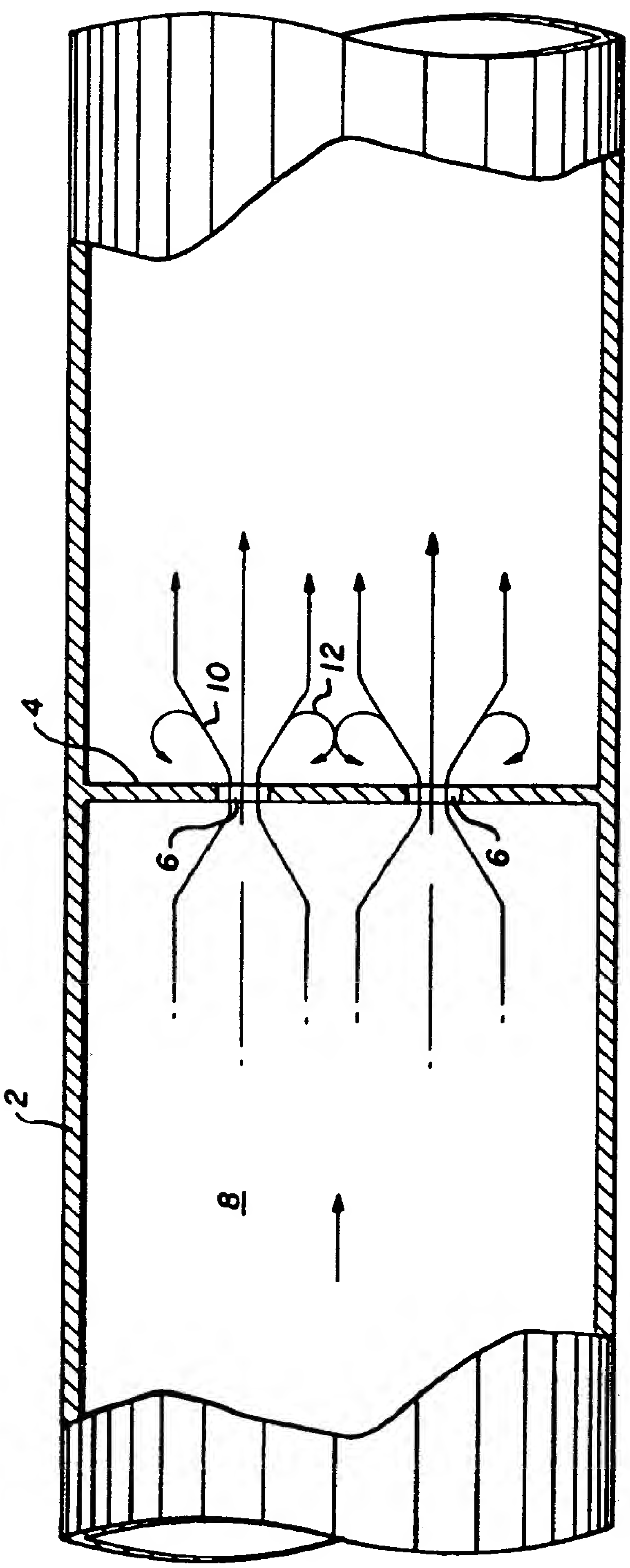


Fig. 1.

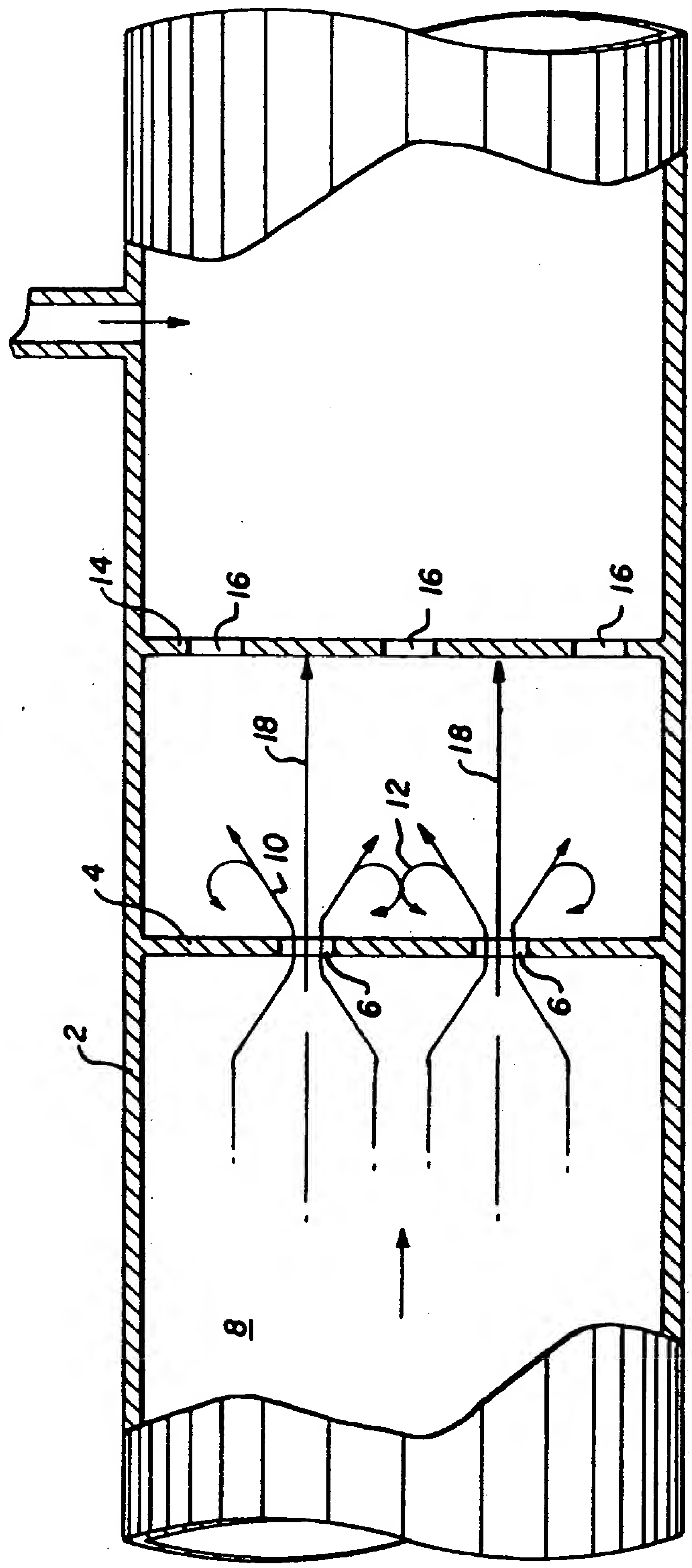


Fig. 2.

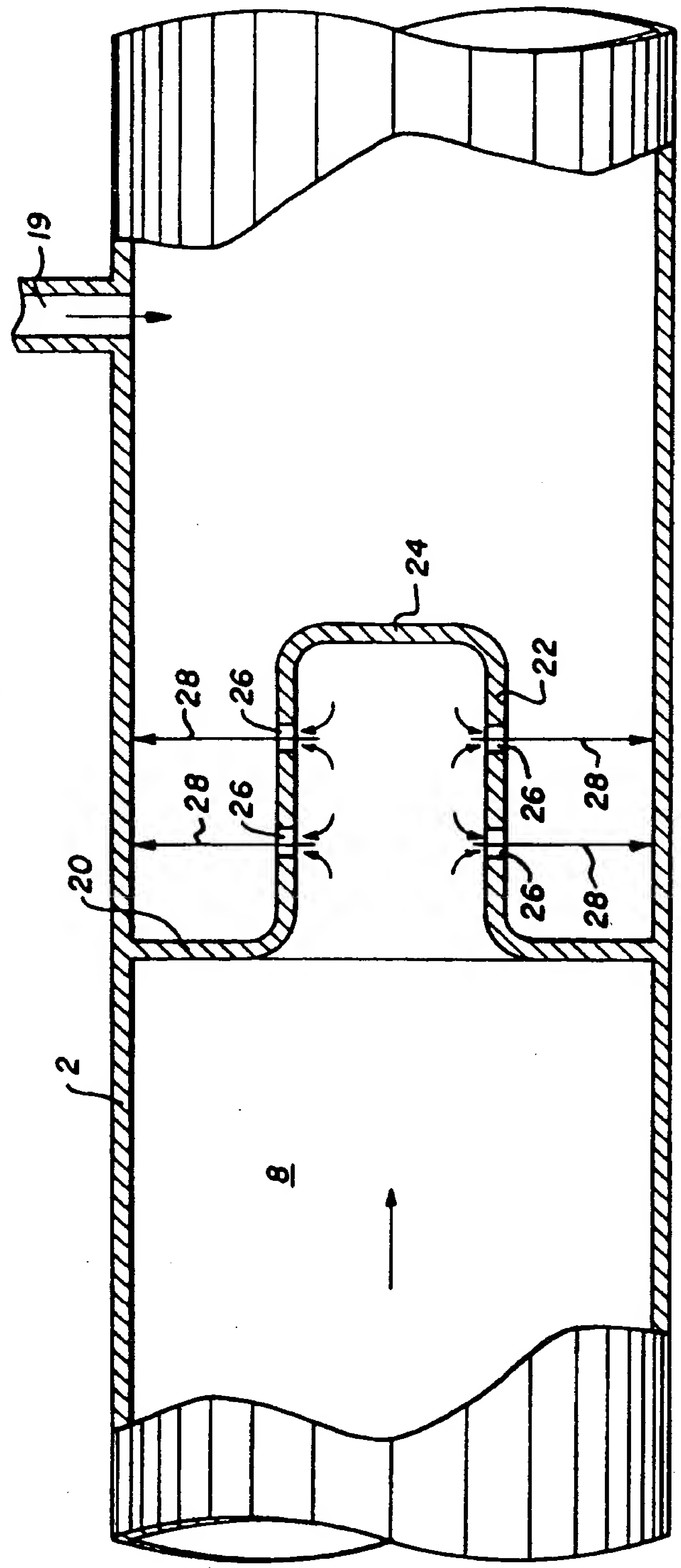


Fig. 3.

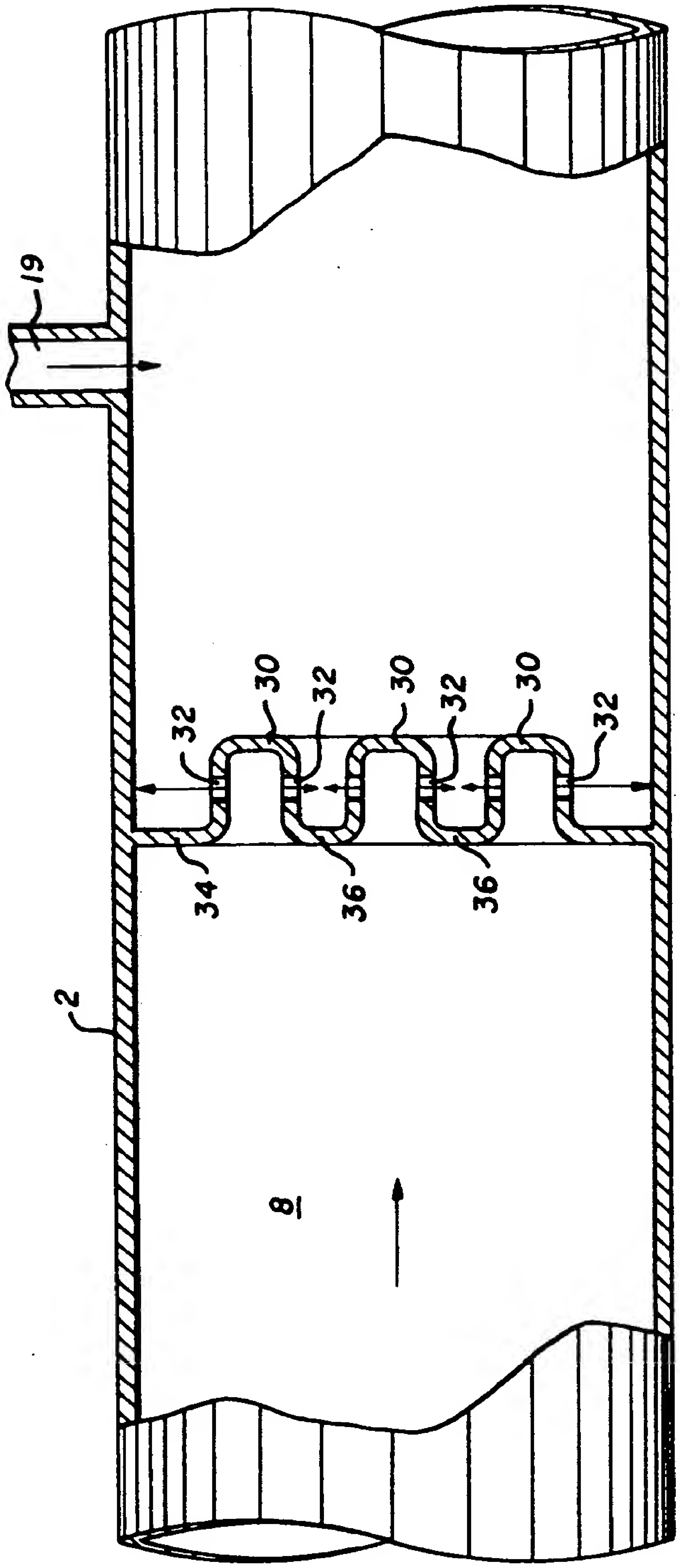


Fig. 4.

Fig. 5.

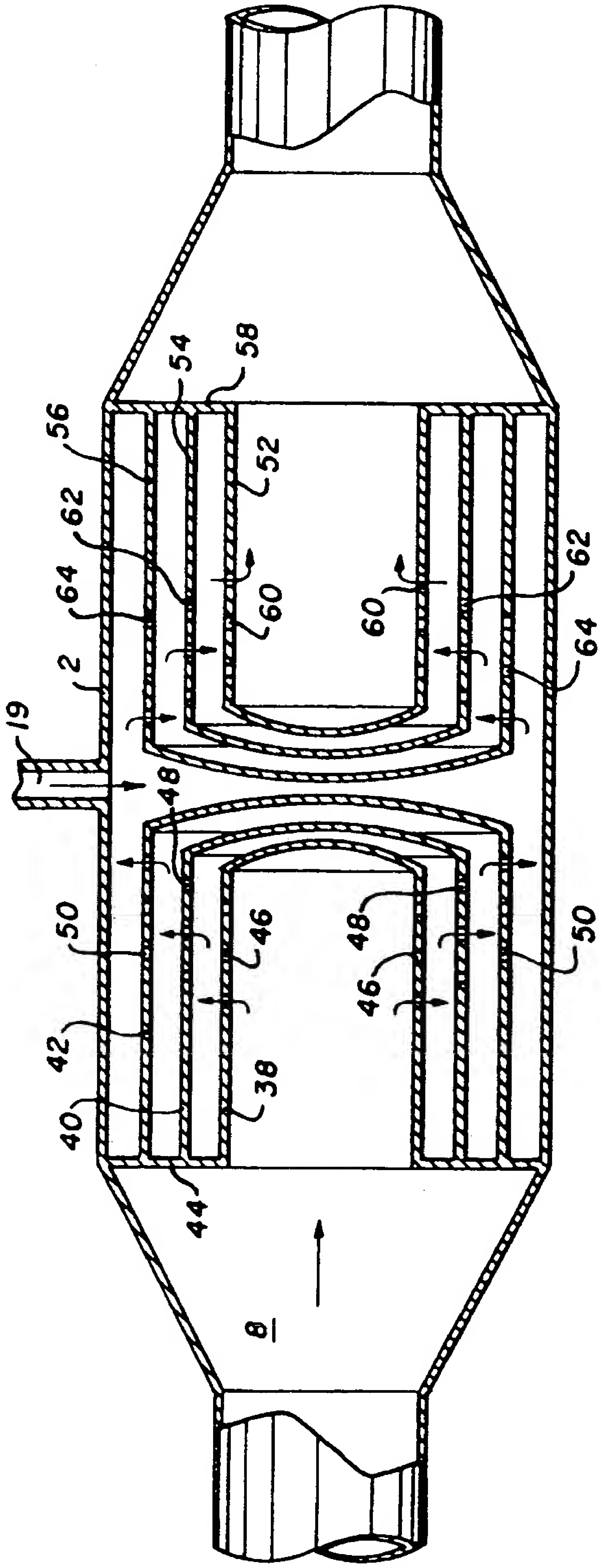


Fig. 6.

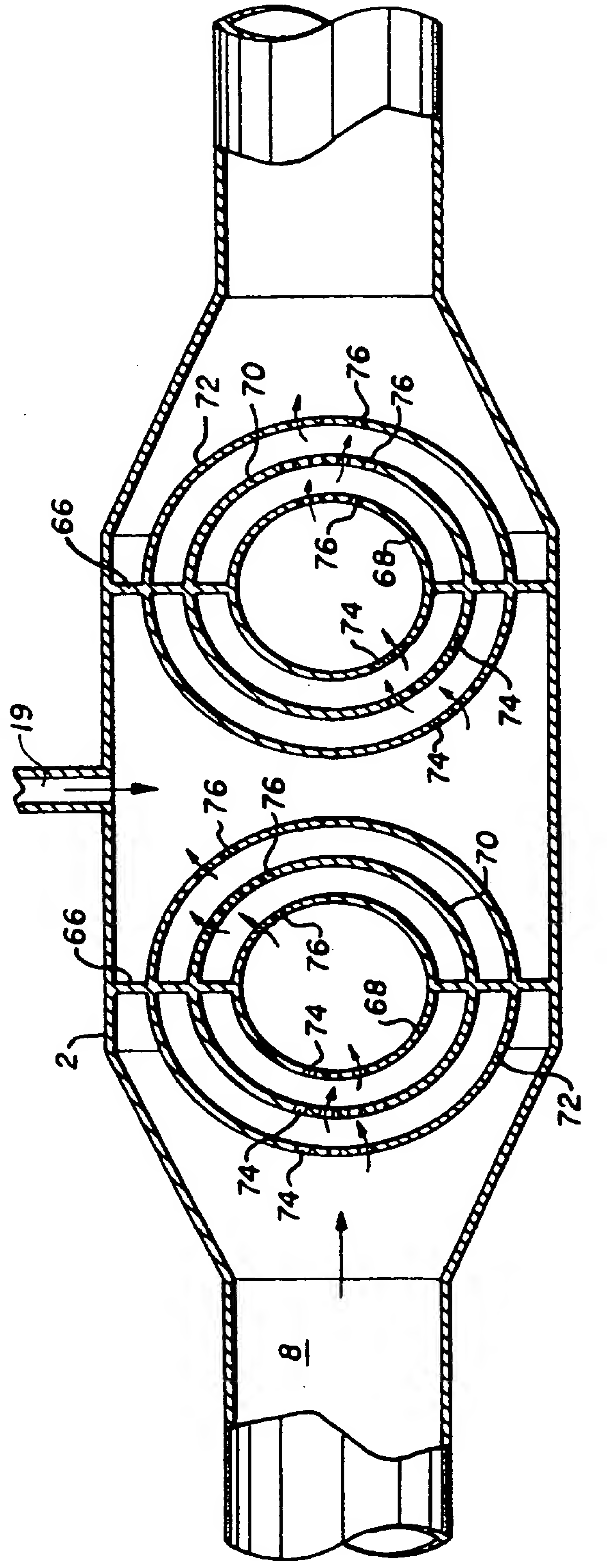


Fig. 8.

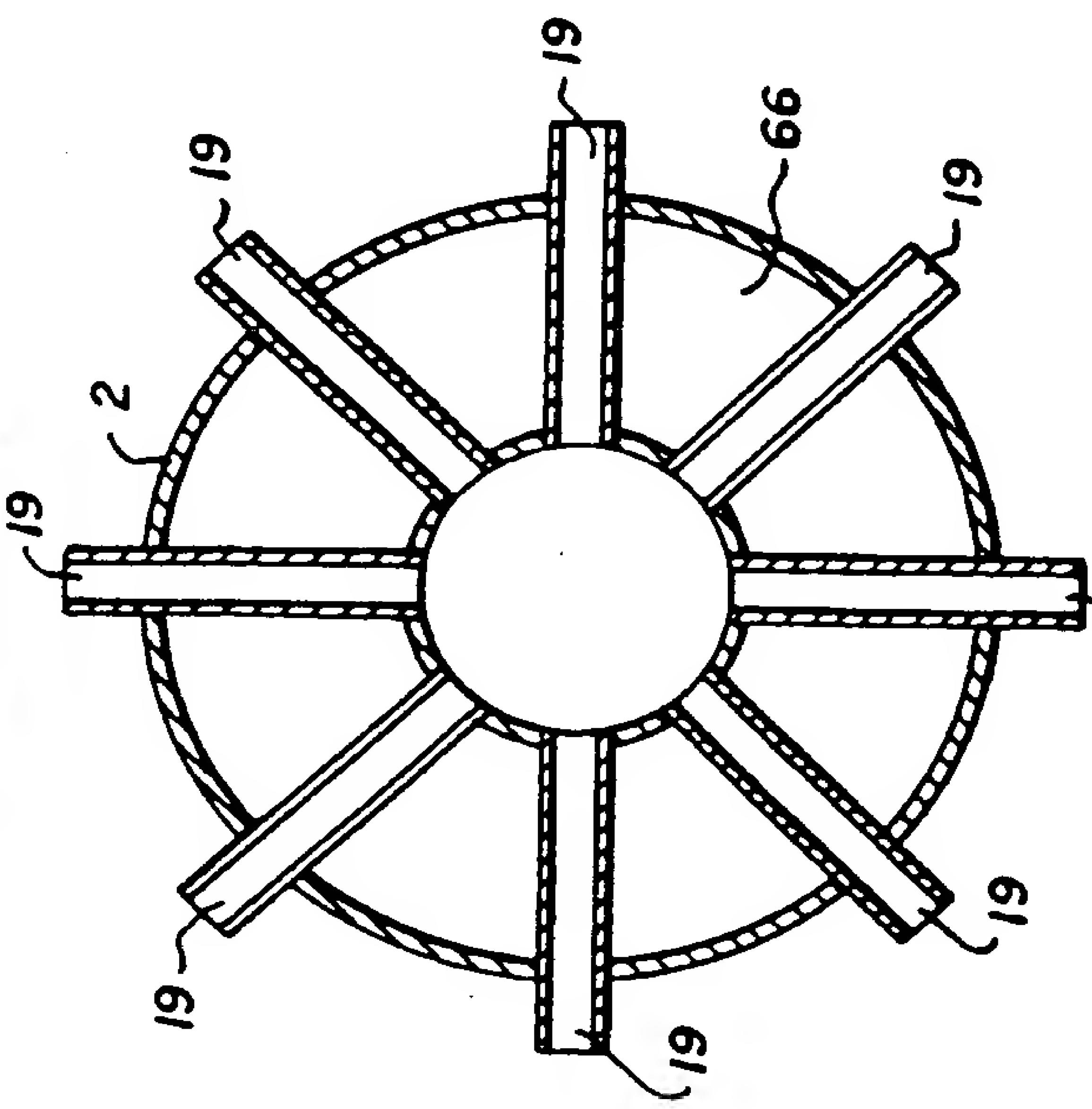
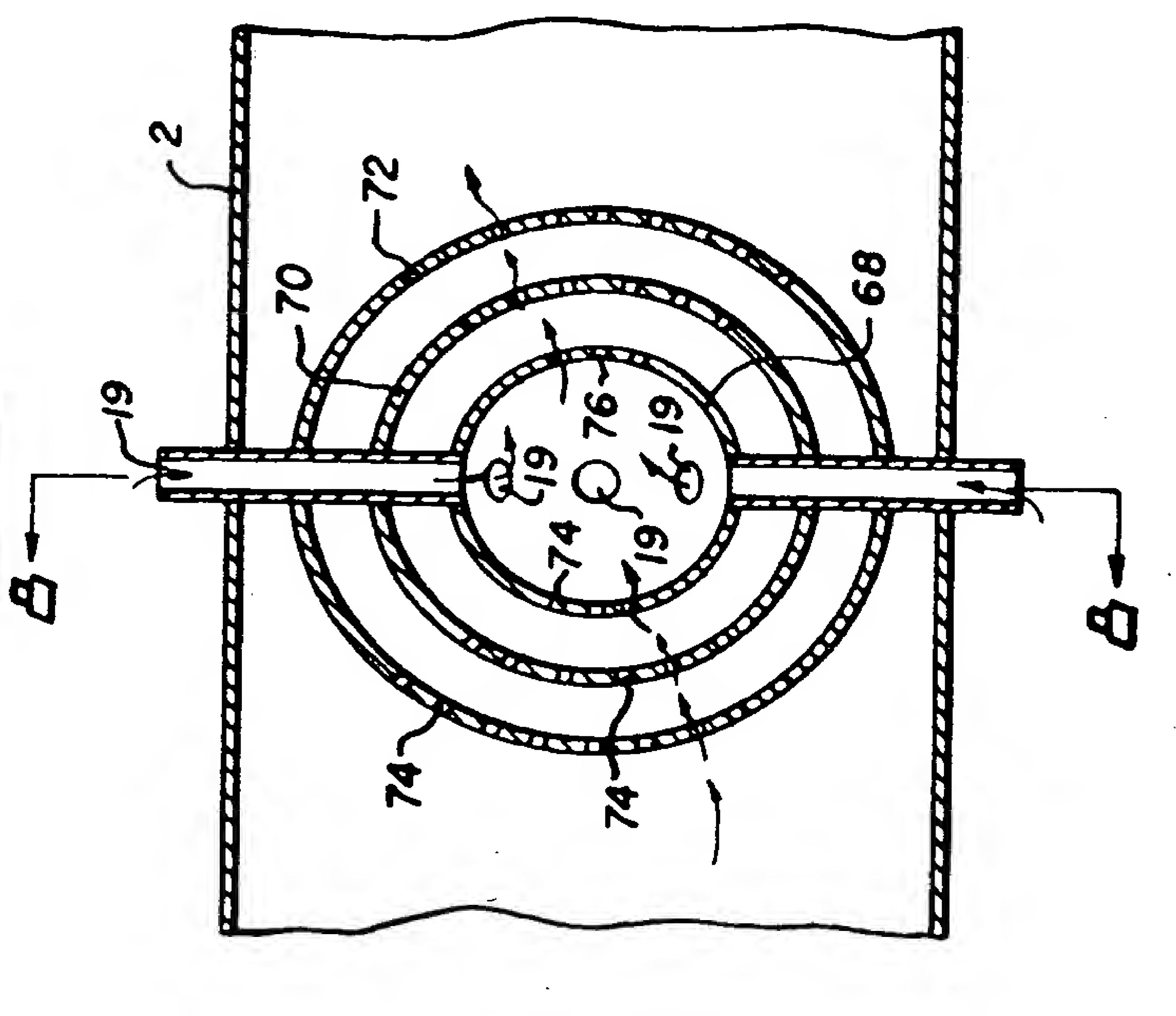


Fig. 7.



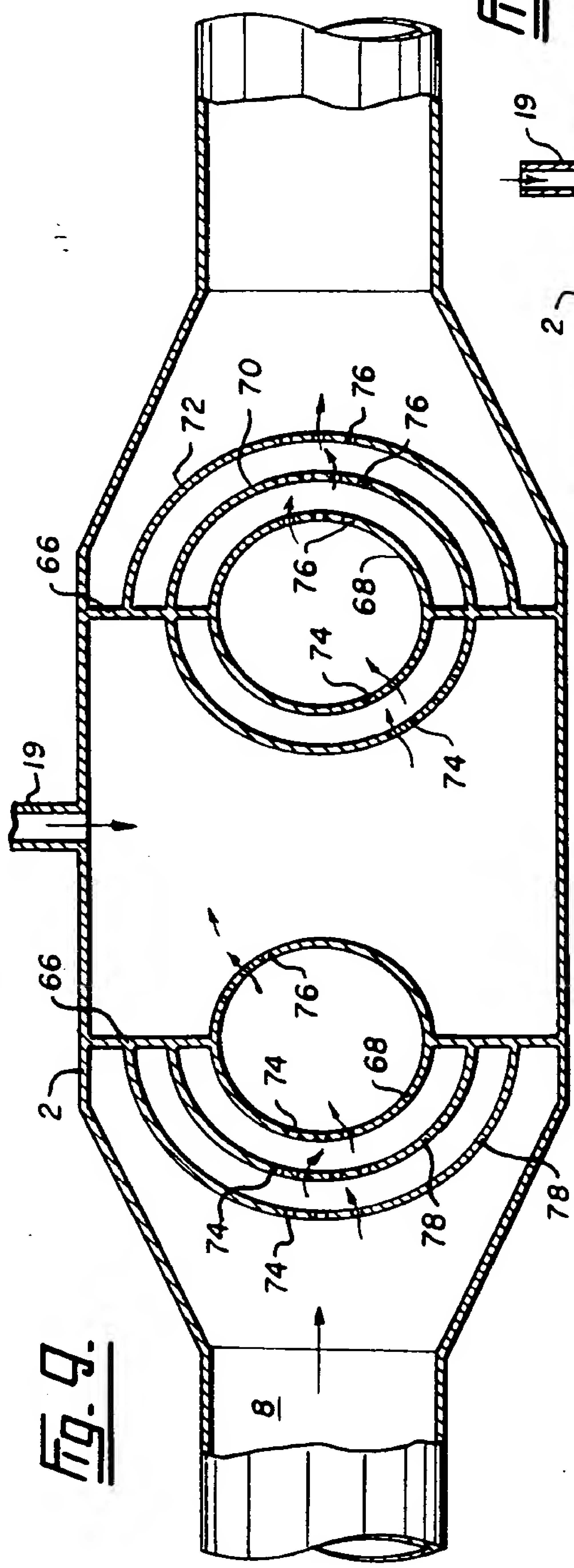

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Fig. 9.

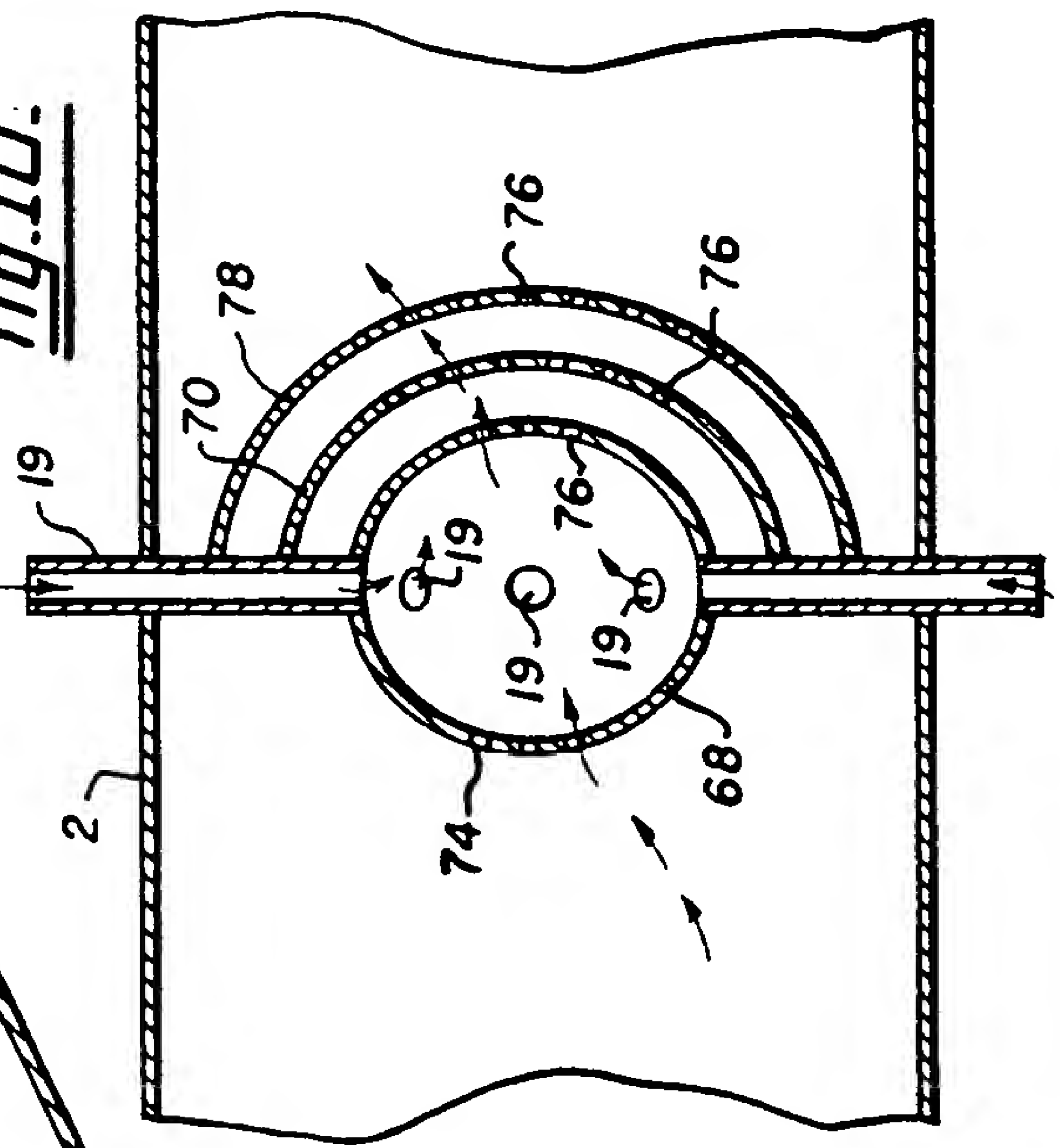
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Fig. 10.

European Patent Office

EUROPEAN SEARCH REPORT

Application Number

EP 90 40 3456

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
X A	US-A-1 698 432 (C.L. ERICKSON) * Column 1, lines 1-10; column 1, line 20 - column 2, line 69; column 2, lines 73-97; figures 1,2 * ---	1,2,5-8,22,25 9,23	B 01 J 14/00 B 01 J 19/26 B 01 J 19/24 B 01 F 5/06
X	PATENT ABSTRACTS OF JAPAN, vol. 7, no. 100 (C-164)[1245], 28th April 1983; & JP-A-58 27 626 (NITSUKOOAAMUZU K.K.) 18-02-1983 * Figures 1-5 * ---	1-4,14,22	
A	DE-B-1 039 049 (NITROGLYCERIN AB) * Column 1, lines 1-7; column 2, line 47 - column 3, line 8; column 4, line 55 - column 5, line 3; column 5, line 38 - column 6, line 8; figure 2 * ---	1,3,4,14,22,25,26	
D,A	US-A-4 043 539 (GILMER et al.) * Abstract; column 1, lines 8-11; column 3, line 62 - column 4, line 56; figures 3,4 * ---	1,2,5-8,12,13,22	TECHNICAL FIELDS SEARCHED (Int. Cl.5)
D,A	US-A-4 136 976 (LEFFELMAN) * Abstract; column 1, lines 5-7; column 1, line 14 - column 2, line 8; figure 1 * ---	1,2,10,12,13,22	B 01 J B 01 F
A	US-A-4 596 699 (DESGRANDCHAMPS et al.) * Abstract; column 1, lines 7-11; column 7, lines 1-40; figure 2 * -----	1-4,12,14,22	

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	02-08-1991	STEVNSBORG N.

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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